Chapter 2. Infrared Nonlinear Optics

Academic and Research Staff

Professor Peter A. Wolff, Dr. Sunny Y.C. Auyang, Dr. Piotra Becla, Professor L. Ramdas Ram-Mohan[1]

Graduate Students

Ady Levy, Cynthia McIntyre, Jason Stark, David B. Walrod, Stephen Wong

2.1 Magnetoresistance of HgMnTe

Sponsor

Defense Advanced Research Projects Agency
Universities Research Initiative
(Contract N00014-46-K-0760)

Project Staff

Dr. Piotr Becla, Simon Foner, Professor Peter A. Wolff, Stephen Wong

At low temperatures, p-type HgMnTe has an enormous negative magnetoresistance.[2] In some cases, magnetic fields of 6T decrease its resistivity by factors as large as 10^5 to 10^6. To determine the mechanism of the large conductivity change, we have performed pulsed, high electric field measurements of current-voltage characteristics in p-HgMnTe. The form of the i-v curves suggests that the large conductivity change results from a magnetic field induced, insulator-metal transition. This conclusion is also consistent with very low temperature transport measurements.[3]

Previous theoretical calculations[4] have shown that modest magnetic fields ($B \simeq 5T$) reorder the valence band states in p-HgMnTe, causing a pancake-shaped acceptor wave function with a large radius in the transverse direction. The insulator-metal transition is then caused by the increased spread of the acceptor state. The theory also implies that the magnetic pressure recompresses the acceptor to a spherical form at much higher fields. This conclusion has been confirmed with magnetoresistance measurements in the Francis Bitter National Magnet Laboratory pulsed field facility.[5] Above 20T, the resistivity of cold p-HgMnTe increases steadily — suggesting an ultimate return to the insulator state — with dramatic changes in the ratio of parallel to perpendicular conductivity.

2.2 Mode Mixing in Narrow Gap Semiconductors

Sponsor

Strategic Defense Initiative/Innovative
Science & Technology, managed by the
U.S. Naval Research Laboratory
(Contract N00014-87-K-2031)

Project Staff

Professor L. Ramdas Ram-Mohan, Richard B. Sohn, Hui Xie, Professor Peter A. Wolff

---

1 Worcester Polytechnic Institute, Worcester, Massachusetts.

2 A. Mycielski and J. Mycielski, *J. Phys. Soc. Japan* 49: Suppl. A807 (1980).

3 T. Wojtowicz, T. Dietl, A. Sawicki, W. Plesiewicz, and J. Jaroszynski, *Phys. Rev. Lett.* 56:2419 (1986).

4 A. Mycielski and J. Mycielski, *J. Phys. Soc. Japan* 49: Suppl. A807 (1980).

5 S.B. Wong, P.A. Wolff, S. Foner, and P. Becla, *Bull APS* 34:7333 (1989), Abstract J15 2; S.B. Wong, *Magnetotransport Studies of the Magnetic Field Induced, Metal-Insulator Transition in $Hg_{1-x}Mn_xTe$*. Ph.D. diss., Dept. of Phys., MIT, 1988.

The effects of interband transitions induced by resonant longitudinal-optic (LO) phonons have been studied theoretically in narrow gap $Pb_{1-x}Sn_xTe$[6] and $Hg_{1-x}Cd_xTe$.[7] With the bandgap, E_g, close to the LO phonon energy, $\hbar\omega_L$, we: 1) evaluate the dielectric function containing contributions from interband transitions induced by the LO phonons and by the Coulomb potential interacting with carriers, 2) show that the LO phonon-intraband plasmon mixing is strongly influenced by the interband transitions, leading to a downward "renormalization" in energy of the mixed mode, and 3) calculate the threshold for stimulated emission of LO phonons when a crystal with $E_G \simeq \hbar\omega_L$ is optically pumped.

These theoretical predictions could be tested with infrared reflectivity experiments. The calculated reflectivity spectra are substantially changed by interband processes. Moreover, the threshold for stimulated phonon emission ($I_{threshold} \simeq 1\,kW/cm^2$) is well within reach of CO_2 laser technology.

2.3 Plasma Instabilities in Quasi-Two-Dimensional Electron Gases

Sponsor

National Science Foundation
(Grant EET-87-18417)

Project Staff

Jason Stark, Professor Peter A. Wolff

Recent pulsed optical experiments in n-doped GaAs quantum wells[8] suggest that hot carriers relax in times as short as 10 fsec, while p-type or undoped quantum wells have relaxation times on the order of 50 fsec. This discrepancy is surprisingly large. While Monte Carlo calculations[9] can account for some of the trends in hot carrier relaxation experiments, times as short as 10 fsec are difficult to understand in terms of single-particle relaxation mechanisms.

As an alternative, we have theoretically studied[10] the possibility of collective mode instabilities in these 2D systems. The dielectric function of a quasi-two-dimensional electron gas is calculated, in the random phase approximation, for a plasma with a non-thermal, excited component. From this function, the plasma eigenfrequencies are determined for the case of a degenerate electron plasma in the presence of electrons excited to energy, E_0, above the Fermi energy. Under such excitation, intrasubband plasma modes experience gain by inverse Landau damping, via coupling to the energetic electrons. This behavior contrasts with that of a three-dimensional plasma, which is stable to the production of plasmons for any spherically symmetric excited distribution.[11] The instability of two-dimensional plasmons is caused by singularities in the one-dimensional density of states for electrons travelling parallel to the plasma mode. The unstable modes experience amplification at rates on the order of the plasma frequency, relaxing energy from the excited distribution. While these growth rates are not fast enough to account for the observed relaxation rates in n-doped quantum wells, the calculations indicate an alternative relaxation path, and imply that two-dimensional plasmas are less stable than those in three dimensions.

To date, these calculations have been restricted to the case of a strictly two-

6 L.R. Ram-Mohan, R.B. Sohn, and P.A. Wolff (to be published).

7 H. Xie, L.R. Ram-Mohan, and P.A. Wolff (to be published).

8 W.H. Knox, D.S. Chemla, G. Livescu, J.E. Cunningham, and J.E. Henry, *Phys. Rev. Lett.* 61:1290 (1988).

9 S.M. Goodnick and P. Lugli, *Phys. Rev. B* 37:2578 (1988).

10 J.B. Stark and P.A. Wolff (to be published).

11 A.I. Akhiezer, I.A. Akhiezer, R.V. Polovin, A.G. Sitenko, and K.N. Stepanov, *Collective Oscillations in a Plasma*, Cambridge, Massachusetts: MIT Press, 1967.

dimensional plasma, without intersubband transitions. We are now investigating the stability of intersubband modes that are known to have large oscillator strengths. Experiments to test these ideas will be performed by Jason Stark in collaboration with Daniel Chemla and Wayne Knox of AT&T Bell Laboratories.

2.4 Nonlinear Optics of Zero Gap Semiconductors

Sponsor

Defense Advanced Research Projects Agency
Universities Research Initiative
(Contract N00014-46-K-0760)
Strategic Defense Initiative/Innovative Science & Technology, managed by the U.S. Naval Research Laboratory
(Contract N00014-87-K-2031)

Project Staff

Dr. Sunny Y.C. Auyang, Professor Peter A. Wolff

In previous work, we have demonstrated that zero gap semiconductors, including HgTe,[12] HgCdTe, and HgMnTe[13] have large third order optical susceptibilities at 10.6μ. For example, in $Hg_{0.84}Cd_{0.16}Te$ with small band gap at 80 K, $\chi^{(3)} = 1.5x10^{-3}$ esu was measured with CO_2 laser, four-wave mixing experiments. Optical non-linearity in these zero gap materials is attributed to interband population modulation, caused by laser-induced carrier temperature fluctuations. Since the carrier temperature relaxes rapidly (5 psec) to that of the lattice, this process has a far higher saturation power than the conventional band filling non-linearity in wider gap materials.

To confirm this hypothesis, we calculated[14] the non-linear coefficient versus intensity in $Hg_{0.84}Cd_{0.16}Te$. At low intensities, the theoretical value, $\chi^{(3)} = 2x10^{-3}$ esu, is in reasonable agreement with the measured $\chi^{(3)} = 1.5x10^{-3}$ esu. When the laser intensity exceeds I=10 kW/cm^2, $\chi^{(3)}$ falls because of the increase in Auger recombination rate caused by the increase in dynamic carrier density. For instance, at I=1 MW/cm^2, the dynamical carrier density is approximately $3x10^{16}$/cc, with a corresponding electron Fermi energy of 60 meV. The large Fermi energy pushes electrons to states of heavier mass, thereby reducing their mass. This effect is quite pronounced in $Hg_{0.84}Cd_{0.16}Te$, which has nearly zero $(\Gamma_8 - \Gamma_6)$ splitting, but less severe in HgMnTe whose $(\Gamma_8 - \Gamma_6)$ splitting is about 0.2 eV. These calculations predict the saturation behavior observed in our four-wave mixing experiments.

2.5 Optical Nonlinearity Due to Resonant Impurity Scattering in HgCdSe:Fe

Sponsor

Defense Advanced Research Projects Agency
Universities Research Initiative
(Contract N00014-46-K-0760)

Project Staff

Dr. Sunny Y.C. Auyang, Professor Peter A. Wolff

Years ago, Kaw[15] suggested that large optical non-linearities could be caused by *energy-dependent* scattering rates of free carriers in semiconductors. It has since been recognized[16] that this mechanism is not effective in the case of "soft" collisions, such as those

---

[12] P.A. Wolff, S.Y. Yuen, K.A. Harris, J.W. Cook, Jr., and J.F. Schetzina, *Appl. Phys. Lett.* 50:1858 (1987).

[13] S.Y. Auyang, P.A. Wolff, K.A. Harris, J.W. Cook, Jr., and J.F. Schetzina, *J. Vac. Sci. Technol.* A6:2693 (1988).

[14] S.Y. Auyang and P.A. Wolff, *JOSA*, in press; P.A. Wolff, S.Y. Yuen, K.A. Harris, J.W. Cook, Jr., and J.F. Schetzina, to be published in *Proceedings SPIE Meeting, Los Angeles*, 1988.

[15] P. Kaw, *Phys. Rev. Lett.* 21:239 (1968).

[16] P.A. Wolff, S.Y. Yuen, and G.A. Thomas, *Solid State Commun.* 60:645 (1986).

due to ionized impurities, but remains valid for abrupt, energy-dependent scatterings. n-HgCdFeSe provides an excellent example of such a scattering system, since Mycielski *et al.*[17] have shown that the Fe^{3+} level forms a sharp scattering resonance in the conduction band. Thus, large optical nonlinearities due to the Kaw mechanism are anticipated. To test this hypothesis, we have performed[18] four-wave mixing experiments on $Hg_{1-x}Cd_xSe$:Fe samples with x=0.23 and x=0.30. In the former, where the Fe $(3d)^6$ level is resonant with the conduction band, the measured $\chi^{(3)}$ is 20 times larger than in the x=0.30 sample with the d-level in the gap. These measurements substantiate the Kaw mechanism; moreover, our theoretical calculations predict a $\chi^{(3)}$ in agreement with the measured value.

2.6 Four-Wave Mixing in Heavily Doped n-GaAs Epilayers

Sponsor

National Science Foundation
(Grant EET-87-18417)

Project Staff

Dr. Sunny Y.C. Auyang, David B. Walrod, Professor Peter A. Wolff

Recent four-wave mixing experiments[19] have shown that the $\chi^{(3)}$'s of heavily doped n-GaAs epilayers are 100 times larger than those of bulk samples with the same doping level. The effect has been observed in a number of n-GaAs epilayers and also in n-InSb epilayers. Since the electron plasma frequencies in these highly doped samples are approaching the 10.6 μ laser frequency, we tentatively attribute the enhancement to plasma resonance effects. Calculations and further experiments to test this hypothesis are in progress.

Publications

MIT Thesis

Wong, S.B., *Magnetotransport Studies of the Magnetic Field Induced, Metal-Insulator Transition in $Hg_{1-x}Mn_xTe$.* Ph.D. diss., Dept. of Physics, MIT, 1988.

---

17 A. Mycielski, P. Dzwonkowski, B. Kowalski, B.A. Orlowski, M. Dobrowolski, and J.M. Baranowski, *J. Phys.* C19:3605 (1986).

18 P.A. Wolff, S.Y. Auyang, R.R. Galazka, and A. Mycielski, *J. Vac. Sci. Technol.* A6:2696 (1988).

19 D.B. Walrod, S.Y. Auyang, P.A. Wolff, and R. Nahory, *Bull. APS* 34:460 (1989), Abstract B147.